

Suitability of Extraction Systems with Macrocyclic Ligands for the Study of Rutherfordium

2. Zr and Hf Extraction with Dibenzo-18-crown-6

R. Sudowe¹, Ch. E. Düllmann^{2,3}, L. M. Farina^{2,3}, C. M. Folden III^{2,3}, K. E. Gregorich², S. E. H. Gallaher^{2,3}, D. C. Hoffman^{2,3}, D. C. Phillips^{2,3}, J. M. Schwantes³, R. E. Wilson^{1,3}, P. M. Zielinski^{2,3} and H. Nitsche^{2,3}

¹ Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

² Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

³ Department of Chemistry, University of California, Berkeley, California 94720-1460

In another contribution to this annual report, we reported on the extraction of zirconium and hafnium with dicyclohexano-18-crown-6 (DC18C6) [1]. In this report the results of a study on the extraction of zirconium and hafnium with dibenzo-18-crown-6 (DB18C6) from various concentrations of hydrochloric acid are presented. These studies are part of an ongoing effort to test the suitability of extraction systems with macrocyclic ligands as model systems for the study of rutherfordium.

The extraction behavior was studied using isotopes with minute half-lives. The distribution ratios for the elements were determined as a function of acid and ligand concentration. The use of DB18C6 for the separation of zirconium and hafnium has been reported previously in the literature [2], but this study was not conducted on a time scale short enough to ensure that the reaction kinetics are fast enough to allow the study of rutherfordium.

The experiments were performed at the 88-inch cyclotron at LBNL using the short-lived isotopes ⁸⁵Zr ($T_{1/2} = 7.9$ m) and ¹⁶⁹Hf ($T_{1/2} = 3.25$ m). The isotopes were produced by bombarding a ^{nat}Ge with an ¹⁸O⁴⁺ beam and a ¹²⁴Sn target with a ⁵⁰Ti¹¹⁺ beam, respectively. The two beams were delivered by the cyclotron as a cocktail beam [3]. The use of this ion cocktail allowed for the fast switching between the two beams. Together with the use of an adjustable target ladder in the Berkeley Gas-filled Separator (BGS) it ensured that experiments with zirconium and hafnium could be performed almost simultaneously and under virtually the same experimental conditions.

The zirconium and hafnium isotopes produced were separated from the beam and interfering reaction products using the BGS. After traveling through the BGS, the products passed through a 3.6 μ m Mylar window into the Recoil Transfer Chamber (RTC) [4]. Inside the RTC, the recoils were thermalized in helium and transported to the chemistry setup using a potassium chloride aerosol gas-jet. The gas-jet was operated with an average flow rate of 1.8 L/min and a pressure of 1.2 bar.

The aerosols containing the radionuclides were transported over a distance of ~15 meters and deposited on platinum foils. The aerosol residue was dissolved in 50 μ L HCl of appropriate concentration and transferred to a centrifuge cone containing additional 3950 μ L of the same acid. The aqueous phase was mixed with an equal volume of crown ether diluted in dichloromethane. Phases were vigorously mixed for 20 seconds and centrifuged for 20 seconds. Afterwards a 3-mL aliquot was taken from each phase and assayed using a HPGe γ -ray detector.

Figure 1 shows the yield for the extraction of Zr and Hf with 0.090 M DB18C6 from 8.0 – 10.5 M HCl. Both elements are extracted, and the extraction yield increases with increasing acid concentration. Zr extraction starts at HCl concentrations above 8.0 M and reaches a maximum of 89%. Hf extraction begins between 8.5 and 9.0 M and reaches a maximum value of 84%. The separation factor, defined as the ratio of extraction yields for the two elements, has its maximum at the lowest acid concentration studied and decreases with increasing acid concentration.

The extraction yield reaches generally lower values than with DC18C6. As with DC18C6, the Hf extraction starts at higher acid concentration compared to Zr.

Both crown ethers investigated show great potential as extractants for the study of the chemical behavior of rutherfordium. They show a stronger tendency to form complexes with Zr than with Hf. Suitable experimental conditions can be chosen to separate Zr from Hf. Using similar conditions when studying the chemistry of rutherfordium should answer the question whether Rf behaves more like Zr or Hf in this chemical system.

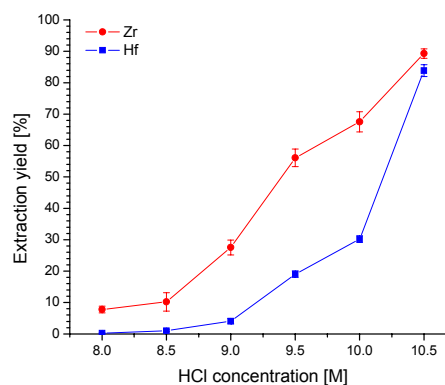


FIG. 1: Zr and Hf extraction from 8.0 – 10.5 M HCl into 0.090 M DB18C6 in CHCl_3 .

REFERENCES

- [1] R. Sudowe et al., contribution to this annual report
- [2] Y. K. Agrawal, S. Sudhakar, Sep. Purif. Technol. **27**, 111 (2002)
- [3] D. Leitner et al., Proc. 15th Int. Workshop on ECR Ion Sources, ECRIS '02, Jyväskylä, June 12-14, 2002 and LBNL-51451 (2002)
- [4] U. W. Kirbach et al., Nucl. Instrum. Meth. A **484**, 587 (2002)